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Polarized photoluminescence excitation spectroscopy of a-plane InGaN/GaN multiple quantum wells grown on r-plane sapphire

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We have performed a detailed study of the impact of basal plane stacking faults (BSFs) on the optical properties of both a-plane InGaN/GaN quantum wells (QWs) and GaN template samples grown on r-sapphire. In particular, we have used polarised photoluminescence excitation spectroscopy (P-PLE) to investigate the nature of the low temperature recombination as well as extracting information on the valence band (VB) polarisation anisotropy. Our low temperature P-PLE results revealed not only excitons associated with intersubband quantum well transitions and the GaN barrier material but also a transition associated with creation of excitons in BSFs. The strength of this BSF transition varied with detection energy across the quantum well emission suggesting that there is a significant contribution to the emission line width from changes in the local electronic environment of the QWs due to interactions with BSFs. Furthermore, we observed a corresponding progressive increase in the VB splitting of the QWs as the detection energy was varied across the quantum well emission spectrum. © *2014 AIP Publishing LLC*.
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I. INTRODUCTION

It is anticipated that the growth of InGaN/GaN based light emitting diodes (LEDs) on non-polar crystallographic planes will increase the efficiency of radiative recombination compared with structures grown on polar planes because the strong electrostatic polarisation fields along the growth direction can be eliminated.[1] Another important characteristic of structures grown on non-polar crystal orientations is the ability to emit polarised light. It is envisaged that polarised light LEDs could be used in backlit liquid crystal displays and save up to 30% of energy compared with conventional systems that use a polarizing film.[2] Successful commercialisation of such LEDs greatly depends on minimising the production costs, thus there is a need to understand the properties of non-polar InGaN/GaN quantum wells (QWs) grown on sapphire substrates compared with structures grown on expensive free-standing GaN. Growth of non-polar GaN on sapphire, however, results in the creation of extended defects[3] such as threading dislocations (TD) and basal plane stacking faults (BSF). These defects impact on the optical properties of InGaN QWs grown on non-polar GaN. Our previous work on nonpolar InGaN QWs grown on sapphire concentrated on the role of BSFs in the emission process, with the main conclusion being that there are two distinct recombination processes from undefected areas of the QWs and regions of the QWs intersected by BSFs.[4,5] Other studies have reported that emission involving BSF related centres can completely dominate the emission properties of m-plane InGaN/GaN QWs grown on SiC.[6] The important role played by BSFs was also demonstrated in a study of a-plane epitaxial lateral overgrowth GaN where the efficient trapping of excitons by BSFs was reported along with evidence of exciton localisation in the BSFs.[7] Nevertheless, much more remain to be done to achieve a detailed understanding of the impact of BSFs on the optical properties of non-polar InGaN/GaN QWs, in particular how BSFs may influence the degree of linear optical polarisation (DLP).

Most of the experimental work on the optical polarisation properties of InGaN QWs has been performed using emission spectroscopy.[8–11] To date, there have been no reports on polarised absorption or excitation spectroscopy studies of non- or semi-polar InGaN QW structures. In this paper, we report on optical measurements using polarised photoluminescence excitation spectroscopy (P-PLE) at low temperatures to shed light on the influence of BSFs on the optical properties of non-polar bulk GaN and InGaN/GaN QWs grown on r-plane sapphire substrates.

II. EXPERIMENTAL DETAILS

The samples studied consist of a GaN template and an InGaN/GaN QW sample on a similar template which were grown on r-plane sapphire substrates by metal-organic vapour phase epitaxy using a 6×2 in. Thomas Swan close-coupled shower-head reactor. For the QW sample, the wells were deposited on a 5.5 μm thick a-plane GaN template which incorporated a single SiN_x interlayer, which has been

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shown to reduce the TD density.[12] Typically, the densities of partial dislocations and BSFs are around $1 \times 10^9\,cm^{-2}$ and $2 \times 10^5\,cm^{-1}$, respectively. The InGaN/GaN QW structure consisted of five 4 nm thick layers of InGaN separated by 7 nm thick GaN barriers. The sample reported on here is one of a series in which the QW growth temperature was varied between 730 and 710 °C in order to obtain different average indium fractions for each individual sample. The indium concentration in the QW sample reported on here is estimated to be 6%. The InGaN QWs were found to be fully strained to the GaN template.

The photoluminescence (PL) measurements on the GaN template sample were performed using excitation from a 5 mW He/Cd laser with light of wavelength 325 nm. The P-PLE studies were carried out on both samples using tuneable excitation from a 300 W Xenon lamp followed by 0.25 m monochromator and a rotatable polariser to select the polarisation of the excitation light. The spectral resolution of the excitation spectra was 1 nm. The samples were mounted in a cryostat such that the c-axis was horizontal while the excitation light was incident close to normal to the surface of the sample and the emitted radiation was collected normal to the plane of the sample, this geometry is illustrated in Figure 1. The depolarized emission from the sample was analysed by a 0.85 m double grating spectrometer and a Peltier-cooled GaAs photomultiplier with a lock-in detector.

III. RESULTS AND DISCUSSION

In Figure 2, P-PLE spectra from the $In_{0.056}Ga_{0.944}N$/GaN QW sample are shown. The peaks at 3.342 eV and 3.356 eV are attributed to the excitation of QW excitons associated with the n = 1 electron and hole sub-bands, labelled $|Y\rangle_{QW}$ and $|Z\rangle_{QW}$, respectively, while the peaks at 3.500 eV ($|Y\rangle_{GaN}$) and 3.517 eV ($|Z\rangle_{GaN}$) are due to the creation of excitons either in the GaN barriers or the GaN template. The assignment of these transitions is consistent with our previous work performed on a-plane GaN/AlGaN QWs.[13,15] We measured similar P-PLE spectra for all the QW samples with different indium concentrations; detailed analysis of the energies of the QW transitions and their polarization properties as a function of indium fraction will be presented elsewhere. Of particular interest is the peak indicated as $U_{|Y\rangle}$ at energy of 3.45 eV. The striking characteristic of this feature is that it is strongly linearly polarised in the $E \perp c$ direction. We consider two

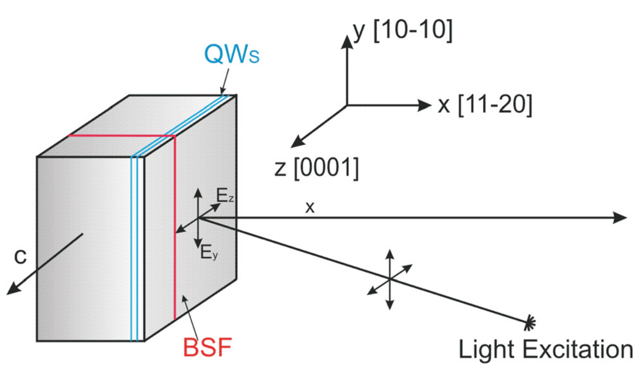

FIG. 1. Experimental geometry of the polarisation dependent P-PLE measurements with the orientation of the QWs and the BSFs as shown.

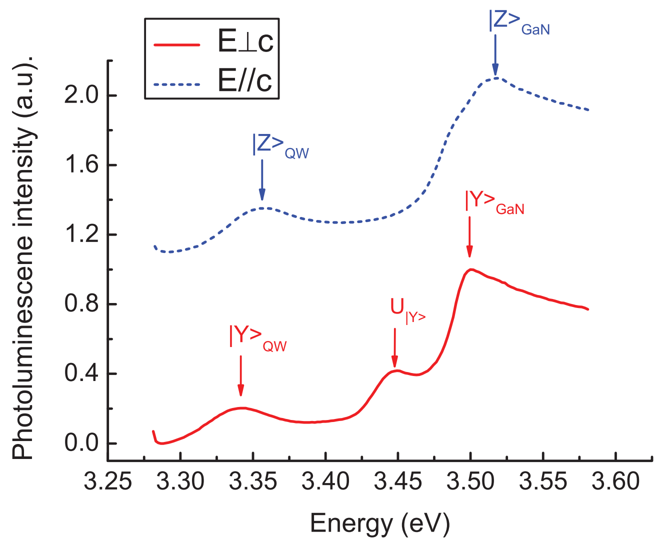

FIG. 2. P-PLE of InGaN/GaN MQW sample recorded at 8 K for the two different polarisations of excitation radiation indicated when detecting at 3.26 eV. The peaks marked $|Y\rangle$ and $|Z\rangle$ represent interband transitions associated with $|Y\rangle$ and $|Z\rangle$ valence sub-bands, respectively.

possible explanations for this transition: (i) a QW transition associated with n > 1 electron or hole sub-bands or (ii) transitions related to BSFs in the GaN. To resolve this situation, we made optical measurements on the GaN template. In Figure 3 are shown PL and P-PLE spectra from the GaN template. The PL spectrum is made up of two strong transitions at 3.494 eV and 3.45 eV which we assign as near band edge (NBE) recombination in the bulk GaN and recombination in the BSFs, respectively.[14] When we perform P-PLE using light with $E \perp c$ polarisation (Figure 3), we observe a transition at 3.45 eV which is spectrally very close to the transition $U_{|Y\rangle}$ in the P-PLE spectrum from the QW sample (Figure 2). Also as for the spectrum shown in Figure 2 the transition at 3.45 eV is not observed for $E \parallel c$. Since the template layer does not contain any QW structure and the fact that optical properties of

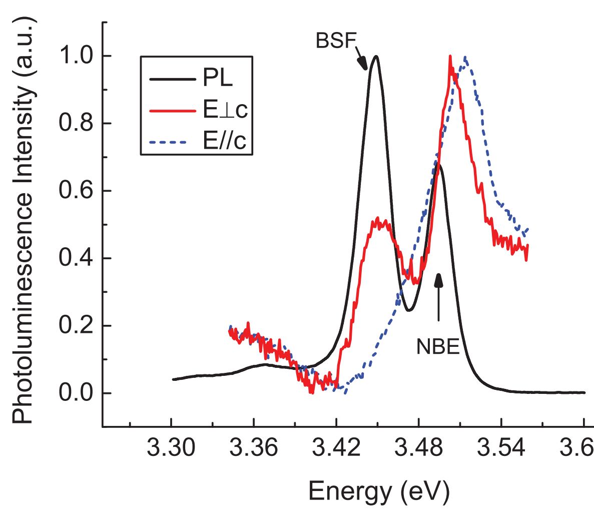

FIG. 3. PL spectrum of GaN template drawn in black solid line with the NBE and BSF emissions as indicated. P-PLE spectra of GaN template sample solid red and dashed blue lines for $E \perp c$ and $E \parallel c$ polarisations, respectively. The detection energy in the P-PLE experiment was set at 2.97 eV. All spectra measured at 8 K.

the $U_{|Y\rangle}$ and transition at 3.45 eV are identical, it allows us to eliminate the possibility that $U_{|Y\rangle}$ involves electron and hole states in the QW with $n > 1$. As the transition at 3.45 eV is very close to the recombination peak from the BSFs (3.448 eV), we initially assign it as being due to the formation of excitons involving the lowest energy valence sub-band in the BSFs. We ascribe the energy difference between the BSF transition in PLE and that in PL as being due to carrier localisation. To add further weight to our identification of $U_{|Y\rangle}$, we now consider the polarisation properties of the P-PLE spectrum in Figure 2. To understand the observed polarisation anisotropy, we must consider the electronic structure of a-plane GaN grown on r-plane sapphire substrates. High resolution X-ray diffraction (HRXRD) measurements have revealed[15] that the growth of GaN on r-sapphire changes the symmetry of the system from being C_{6v} (hexagonal) to C_{2v} (orthorhombic) as well as giving insight into the strain state. The GaN is compressively strained in the y-z plane ($\epsilon_{yy} < 0$; $\epsilon_{zz} < 0$) and is tensile strained along the x-axis ($\epsilon_{xx} > 0$). Here, ϵ_{ii} denotes the diagonal components of the strain tensor. As discussed in detail in Ref. 15, such an anisotropic strain ($\epsilon_{xx} \neq \epsilon_{yy}$) breaks the symmetry between the otherwise degenerate $|X\rangle$ and $|Y\rangle$-like valence band (VB) states and shifts the $|Y\rangle$-like ($|X\rangle$-like) state to higher (lower) energies. It is important to note that the $|Z\rangle$-like VB state is energetically separated from the $|X\rangle$ and $|Y\rangle$-like states due to the crystal field splitting. Based on the experimental geometry shown in Figure 1, we identify the two transitions at 3.342 eV and 3.356 eV as involving the $|Y\rangle$ and $|Z\rangle$-like sub-valence bands, similar to our previous findings on a-plane GaN/AlGaN QWs.

Now we turn our attention again to the feature $U_{|Y\rangle}$ at 3.45 eV in the P-PLE spectrum that we initially suggested was due to excitation of excitons in the BSFs. BSFs can, in principle, be considered as 10 Å wide (111)-oriented ZB inclusions in the wurtzite (WZ) GaN host matrix.[16] Therefore, BSFs can be treated as very narrow QWs in the GaN templates; such that the plane of the BSF QWs is normal to the growth plane as illustrated in Figure 1. Different VB alignments for ZB inclusions in WZ material have been reported in the literature.[17–19] Based on density functional theory (DFT), Murayama and Nakayama[17] and later Stampfl and Van de Walle[18] reported that planar (111)-oriented ZB inclusions in the WZ phase form shallow type-II QWs, with confinement for the electrons in the ZB phase. More recently, however, Belabbes and co-workers,[19] using quasi-particle DFT calculations, came to the conclusion that BSFs in GaN behave as shallow type-I QWs, therefore, confining both electrons and holes in the ZB region.

As we stated earlier, if we consider BSFs as narrow (10 Å width) type I QWs, there will be very strong confinement for holes along the z direction as specified in Figure 1. Consequently, since the $|Z\rangle$-like state has a very low effective mass, almost 10 times smaller than the $|Y\rangle$-like state along this direction,[15] we anticipate a large energy difference between transitions involving the $|Y\rangle$ and $|Z\rangle$ valence sub bands, in fact much larger that the splitting of the VB states detected in the P-PLE spectra shown in Figure 3 for the bulk GaN. On this basis, we extend our assignment of $U_{|Y\rangle}$ as involving the $|Y\rangle$ valence sub-band in the BSFs. The anticipated large splitting is supported by the fact that the feature $U_{|Y\rangle}$ is only visible in the E ⊥ c configuration but not in E || c. If the $|Z\rangle$-like valence sub-band state was energetically close, the spin-orbit interaction would mix $|Y\rangle$- and $|Z\rangle$-like states and a feature near $U_{|Y\rangle}$ should also be visible in E || c configuration. This then begs the question as to where is the transition associated with the $|Z\rangle$-like valence sub-band in the E || c excitation configuration. We note in Figure 3 that the exciton transition in the E || c associated with the GaN 3.517 eV is very much broader that that at 3.500 eV so we suggest that the peak in E || c at 3.517 eV is made up of overlapping contributions from transitions involving the $|Z\rangle$-like states in the bulk GaN and the BSFs. We also note that $U_{|Y\rangle}$ is of comparable strength to that observed for the bulk GaN transitions, which supports the model predicting that BSFs are type I QWs, although the strength of any transition in a PLE spectrum is inevitably governed by other processes specific to the experiment in question, such as carrier transfer processes.

Following on from our identification of the feature $U_{|Y\rangle}$ at an energy of 3.45 eV in the P-PLE spectrum from the GaN template as being due to an exciton transition associated with BSFs, we turn our attention to the P-PLE spectra in Figure 4 obtained while monitoring the emission from the InGaN QWs. The appearance of $U_{|Y\rangle}$ in the P-PLE spectrum for E ⊥ c indicates that there is a transfer of carriers to the InGaN QWs from the BSFs. We note that as the detection energy is reduced $U_{|Y\rangle}$ increases in strength, and the QW transitions along with those associated with the GaN barriers broaden. This argument is reinforced by the observation that when $U_{|Y\rangle}$ is particularly strong there is also a distinct shoulder on the low energy side of the GaN transitions for E || c compatible with our previous argument that a transition associated with the $|Z\rangle$-like states in the BSFs overlaps with the bulk GaN transitions. We explain the increased strength of the BSF transitions while monitoring the QW emission as follows. In the limit of carriers excited in the GaN barriers

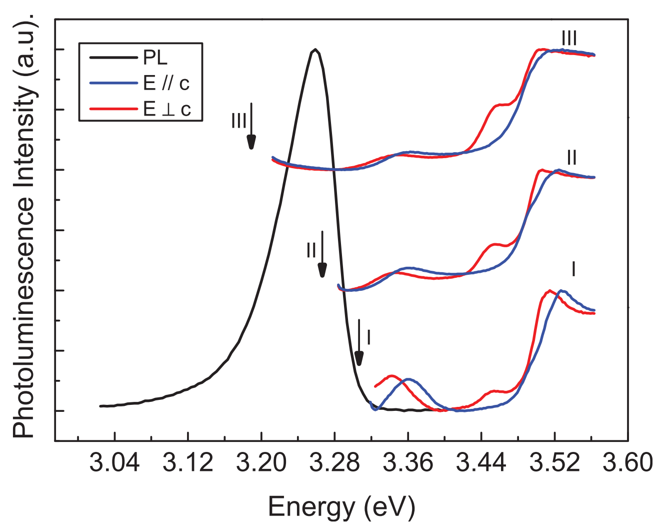

FIG. 4. Photoluminescence spectrum from the MQW sample (black line) and P-PLE for InGaN/GaN MQW at the indicated detection energies I, II, and III corresponding to 3.30, 3.26, and 3.20 eV, respectively. The PLE spectra are offset on the same graph for clarity, the solid red and solid blue lines are when the light was polarised with E ⊥ c and E || c, respectively. All spectra measured at 8 K.

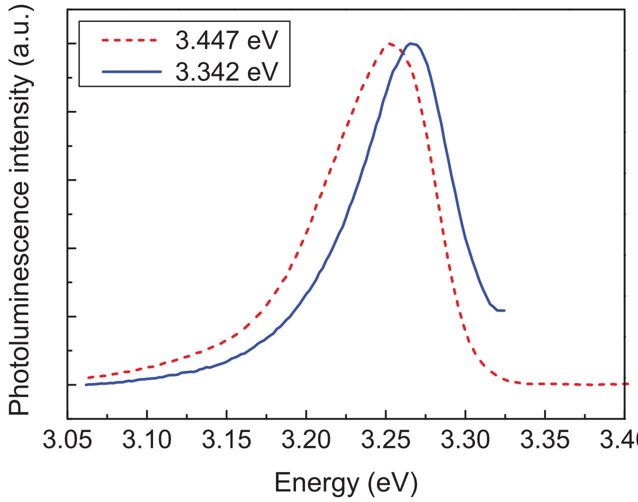

FIG. 5. Photoluminescence spectra measured at a temperature of at 8 K. The solid blue line and dashed red line are for spectra excited by light of excitation photon energy of 3.342 eV and 3.447 eV, respectively. The excitation light was polarised with $E \perp c$.

far away from a BSF, we expect to see sharp transitions associated with excitation of excitons in both the GaN barriers and the QWs. We come closest to this scenario when we monitor light from the high energy side of the PL spectrum. As the detection energy is progressively reduced the BSF transition becomes increasingly dominant. As carrier diffusion lengths in the plane of the InGaN QW are likely to be restricted by localisation effects then we interpret the increasing strength of the BSF transition as the detection energy is decreased as reflecting the fact that we monitor the recombination of carriers exited close to BSFs. This view is compatible with the broadening of the GaN transitions and the QW transitions caused by the local strain environment close by the BSFs. This overall behaviour is reflected in selective excitation spectra shown in Figure 5. For these two spectra, an excitation photon energy of 3.45 eV is used which leads to excitation of the BSF affected emission or 3.342 eV where carriers are directly excited into the QWs. As expected, on the basis of the discussion of the P-PLE spectra above, the peak emission shifts to lower energy when the BSFs are excited.

IV. CONCLUSION

We have performed detailed P-PLE spectroscopy of the recombination from BSFs in a-plane InGaN/GaN QWs and a GaN template sample grown on r-sapphire. From P-PLE measurements on the template sample, we identified the strongly polarised feature $U_{|Y\rangle}$ attributed to the creation of excitons confined at the BSFs. Based on this assignment, we showed that the emission from the low energy side of the PL spectrum from an InGaN/GaN QW is perturbed by the presence of BSFs. Furthermore, our experimental studies hint that BSFs are narrow type-I QWs, in line with recent theoretical predictions. Thus BSFs make a clear contribution to the inhomogeneously broadened PL spectrum from a-plane InGaN/GaN QWs at low temperatures.

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